LA-UR-21-25821

Title: Electronic Correlation and Topology in f-Electron Quantum Matter

Author(s): Zhu, Jianxin

Intended for: IMS Student Lecture Series

Issued: 2021-06-21

Electronic Correlation and Topology in f-Electron Quantum Matter

Jian-Xin Zhu
Los Alamos National Laboratory

IMS Student Lecture Series, LANL, June 22, 2021

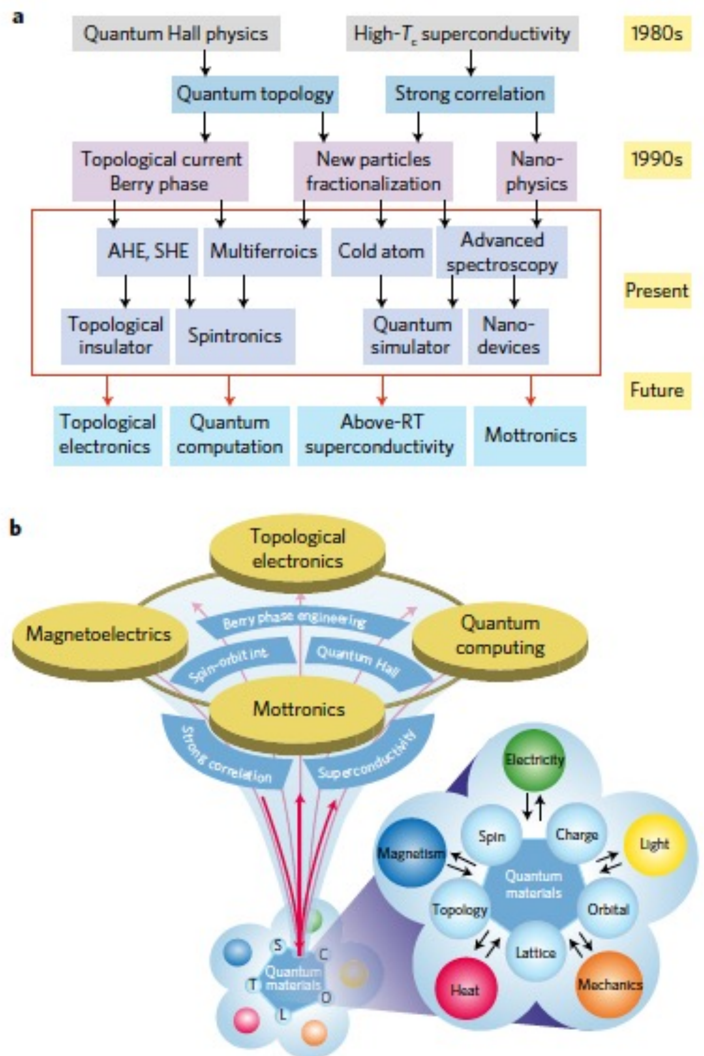
Outline

- Introduction
  - quantum matter
  - topological insulators
  - Interacting topological systems

- f-electron quantum matter
  - Electronic structure and topological classification of PuB_4
  - Electronic structure of $Ce_3Pt_3Bi_4$/$Ce_3Pd_3Bi_4$ family of heavy fermion systems
  - Electronic structure of CeBi

- Summary

Quantum Materials Matter



Tokura, Kawasaki, Nagaosa, Nature Phys. **13**, 1056 (2017)

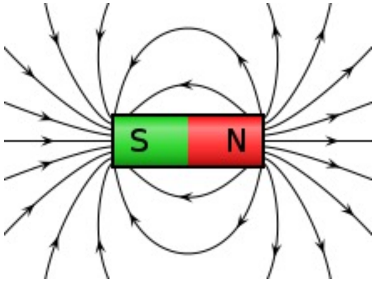
- Before 1980’s, condensed matter systems describable by Landau-Ginzburg paradigm: **symmetry breaking** and **order parameter**
  - Magnetism
  - Conventional superconductivity/superfluidity
- Starting from strongly correlated electron systems in 1980’s, the concept of QMs has now a broader scope, described directly by **electron wavefunction** and **topology**
  - High-temperature superconductors
  - Quantum Hall effect systems
  - Topological insulators
  - Weyl semimetals
  - Quantum spin liquids
  - Graphene
  - 2D Transition-metal dichalcogenides
  - …
- QMs with plethora **degrees of freedom** and **functionality**
  - Test ground of fundamental physics (Dirac fermions, Weyl fermions, Majorana fermions)
  - huge potential for technological applications

In what ways matter can become ordered?

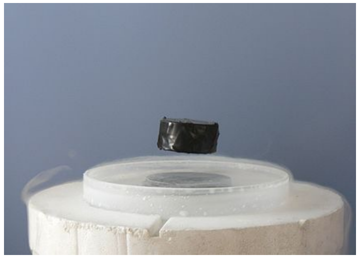
- Landau paradigm

Crystals

Magnets



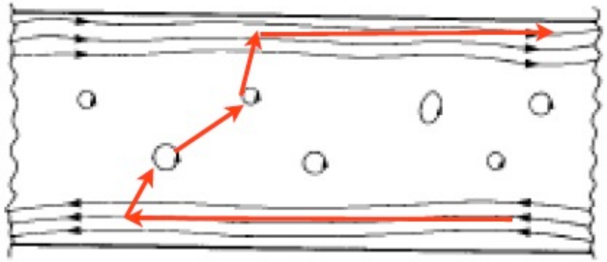
Superconductors



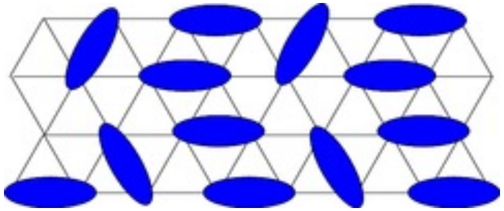
. . . .

- Beyond Landau paradigm

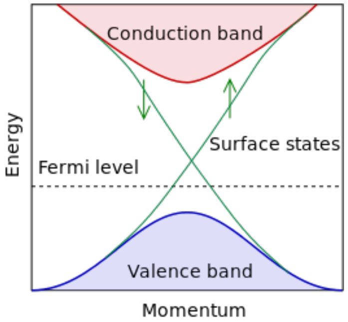
Quantum Hall States



Quantum Spin Liquids

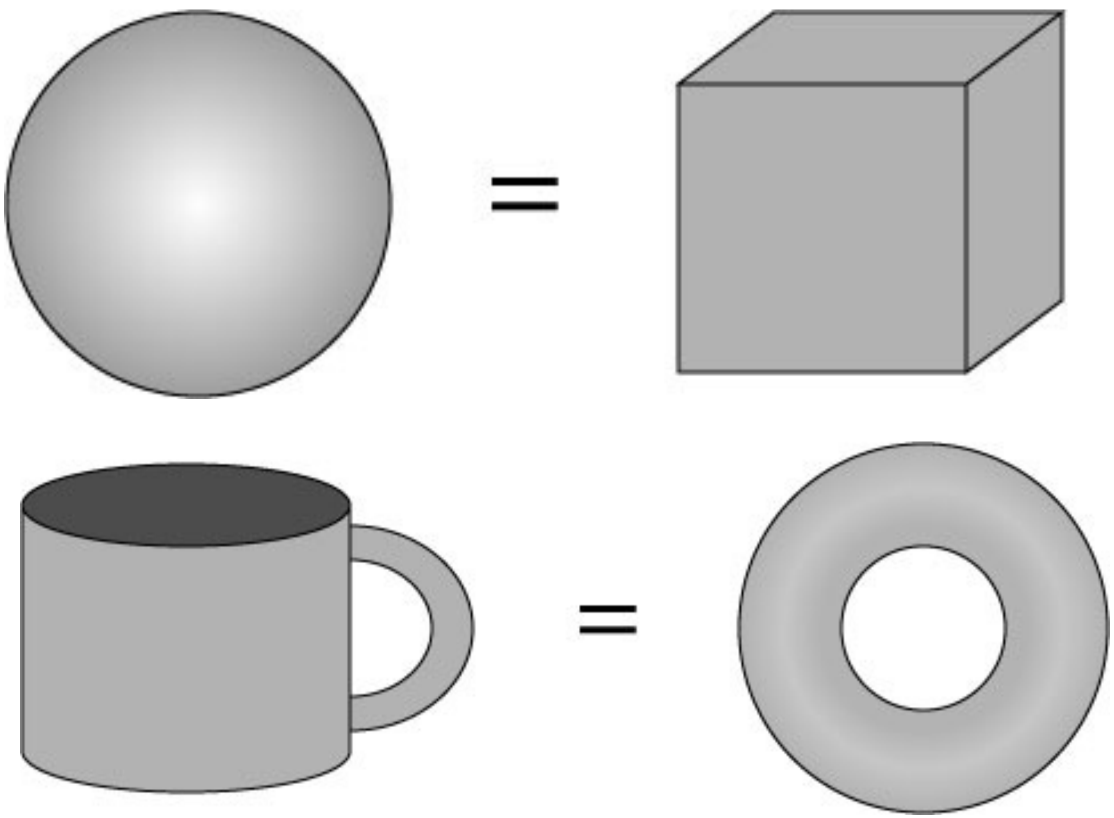


Topological Insulators



Topology

- Branch of mathematics concerned with geometrical properties of objects
- Integer topological invariant called the genus g, essentially the number of holes



- Objects with the same g number are topologically equivalent

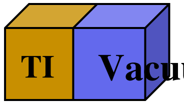
Towards Topological Insulators

a. Insulator
:band gap

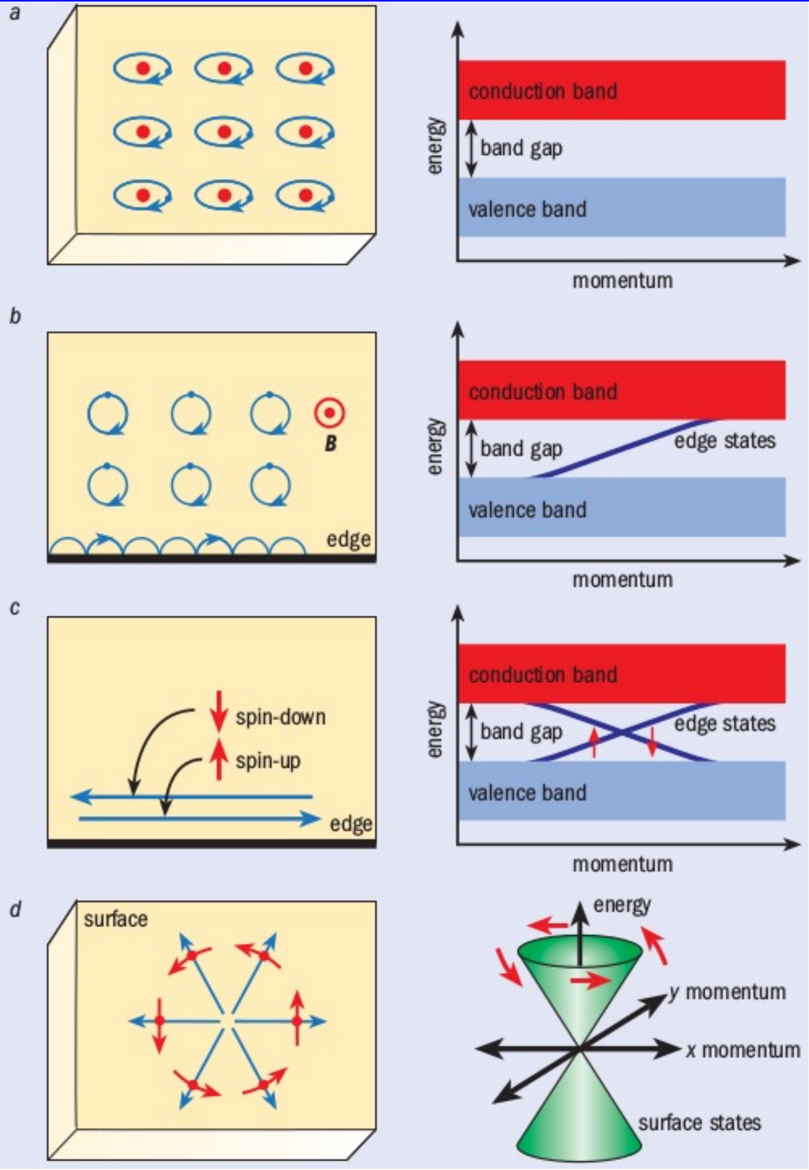
b. Integer quantum Hall effect
:Strong magnetic field, Chern number (TKNN) (C not equal to 0)

c. Spin quantum Hall effect (C=0)
:Time-reversal symmetry (w/ SOC), Z_2 invariant

d. Topological insulator in 3D (C=0)
: four Z_2 invariant indices



Vacuum is trivial insulator!

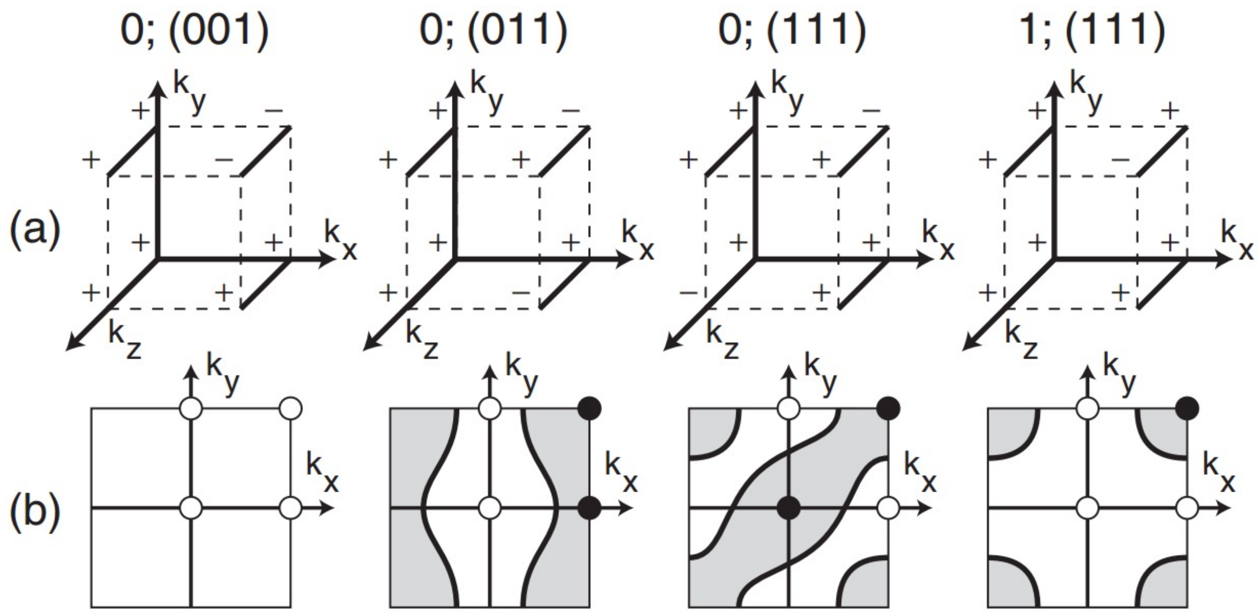


C. L. Kane and J. E. Moore, Phys. World **24**, 32 (2011)

3D Topological insulators

- Four Z_2 invariants needed to identify the 3D TIs

$$PH(k)P^{-1} = H(-k),$$
$$E_m(-k) = E_m(k)$$
$$Pk_0 = k_0, k_0 = 0, \mathrm{mod}(G) + \pi$$
$$P\psi_m(k_0) = \varsigma_m(k_0)\psi_m(k_0),$$
$$\varsigma_m(k_0) = \pm 1$$



$$\Gamma_{i=(n_1 n_2 n_3)} = (n_1 \mathbf{b}_1 + n_2 \mathbf{b}_2 + n_3 \mathbf{b}_3)/2$$

8 time-reversal invariant momentum

$$(-1)^{\nu_0} = \prod_{n_j=0,1} \delta_{n_1 n_2 n_3},$$

$$\Rightarrow \quad \nu_0; (\nu_1 \nu_2 \nu_3)$$

$$(-1)^{\nu_{i=1,2,3}} = \prod_{n_{j\neq i}=0,1; n_i=1} \delta_{n_1 n_2 n_3}.$$

Fu, Kane, and Mele, Phys. Rev. Lett. **98**, 106803 (2007)

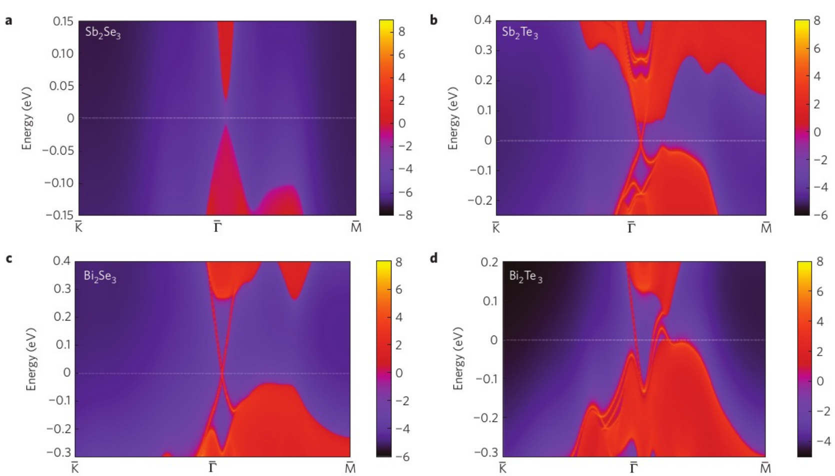
Prediction and Discovery of 3D TIs

- Parity analysis at the symmetry k-point,

14 occupied bands; 1 unoccupied band

Sb_2Se_3	$+-+-+-+-+-+---;+$	$(+)$
Sb_2Te_3	$+-+-++-+-+---+;-$	$(-)$
Bi_2Se_3	$+-+-+-+-+-+--+;-$	$(-)$
Bi_2Te_3	$+-+-+-++-+---+;-$	$(-)$

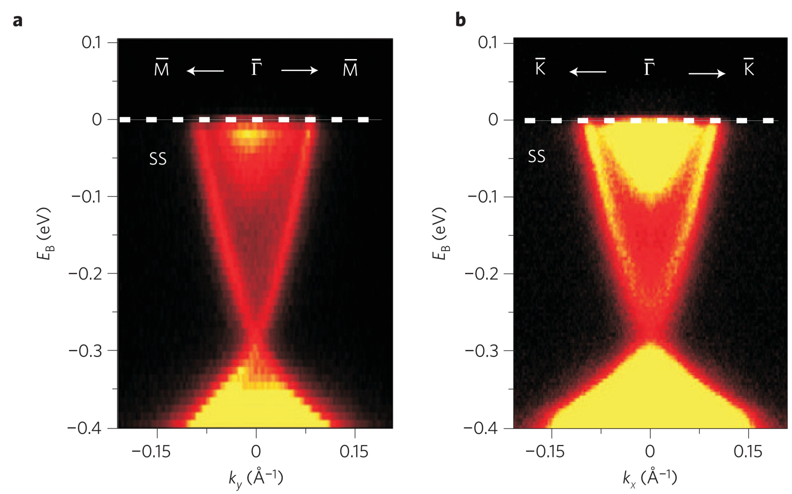
- Slab band structures



Zhang *et al.* Nat. Phys. **5**, 438 (2009)

❖ DFT calculations analyzed topological classification and surface band structure

❖ Angle-resolved photoemission measurement observed the Dirac cone in Bi_2Se_3.



Xia *et al.*, Nature. Phys. **5**, 398 (2009)

- Band topological insulators are important class of quantum materials with the existence of protected surfaces states. Popular!

?Correlation

- **Topological Kondo insulators**
- **Quantum spin liquid**

f-electron

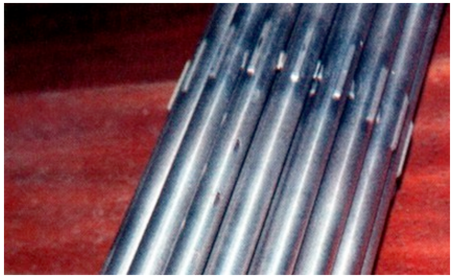
They are rare!

57 La lanthanum 138.91	58 Ce cerium 140.12	59 Pr praseodymium 140.91	60 Nd neodymium 144.24	61 Pm promethium	62 Sm samarium 150.36(2)	63 Eu europium 151.96	64 Gd gadolinium 157.25(3)	65 Tb terbium 158.93	66 Dy dysprosium 162.50	67 Ho holmium 164.93	68 Er erbium 167.26	69 Tm thulium 168.93	70 Yb ytterbium 173.05	71 Lu lutetium 174.97
89 Ac actinium	90 Th thorium 232.04	91 Pa protactinium 231.04	92 U uranium 238.03	93 Np neptunium	94 Pu plutonium	95 Am americium	96 Cm curium	97 Bk berkelium	98 Cf californium	99 Es einsteinium	100 Fm fermium	101 Md mendelevium	102 No nobelium	103 Lr lawrencium

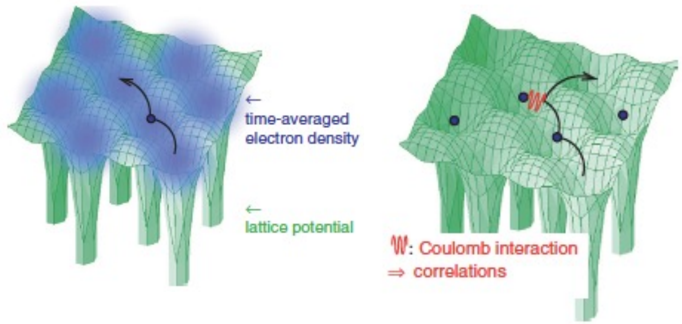
f-electron quantum materials are functional materials

- *d*-electron materials
  - Cuprates, e.g., YBCO (superconductivity -- energy efficiency);
  - Manganites, e.g., LaCaMnO (magnetoresistivity -- information technology)

- *f*-electron materials
  - lanthanides, e.g., $CeRhCo_5$ (heavy fermion superconductivity -- energy efficiency);
  - Actinides, e.g., UO_2 (nuclear fuel rod -- energy generation), $PuCoGa_5$ (heavy fermion superconductivity -- energy efficiency).



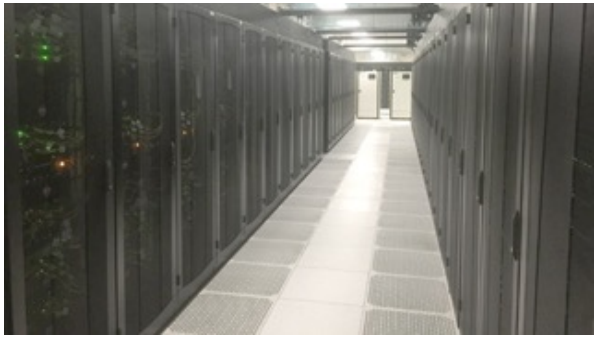
Co-design approach to f-electron quantum materials



Controlled synthesis/fabrication (CINT/MPA-Q)

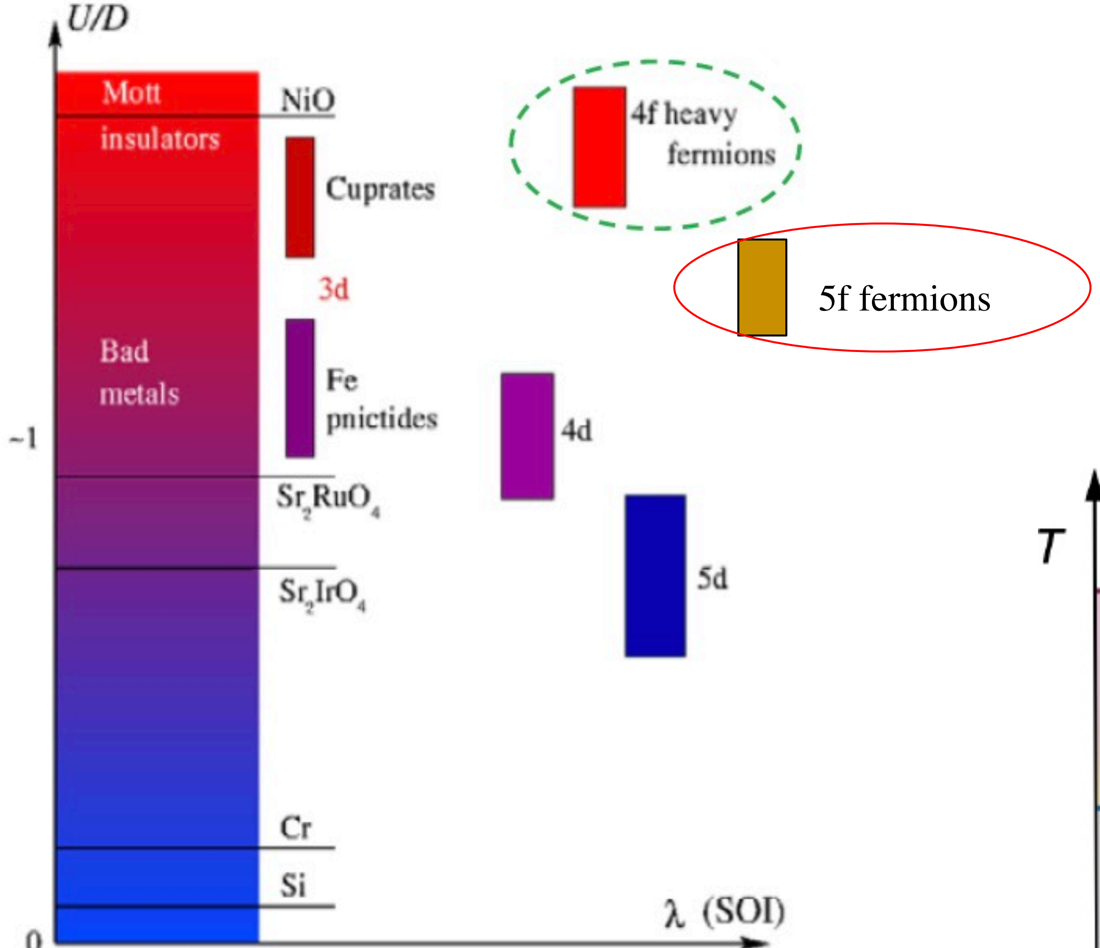
Electronic Structure Theory/Simulation (T)

Characteriztion (CINT/MPA-Q/NHMFL)

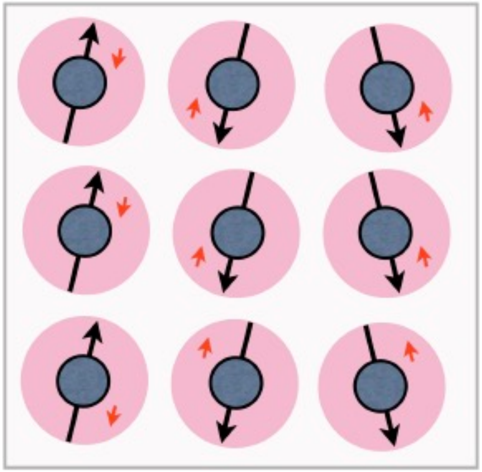


LANL HPC-Institutional Computing

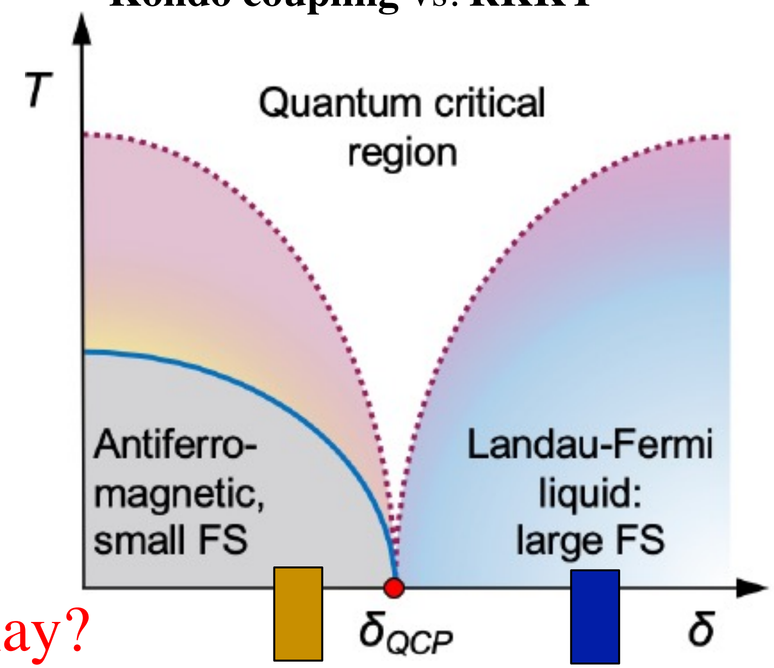
Heavy fermion systems



Courtesy of Sarah Grefe



Kondo coupling vs. **RKKY**



- With SOC, topology and interplay?

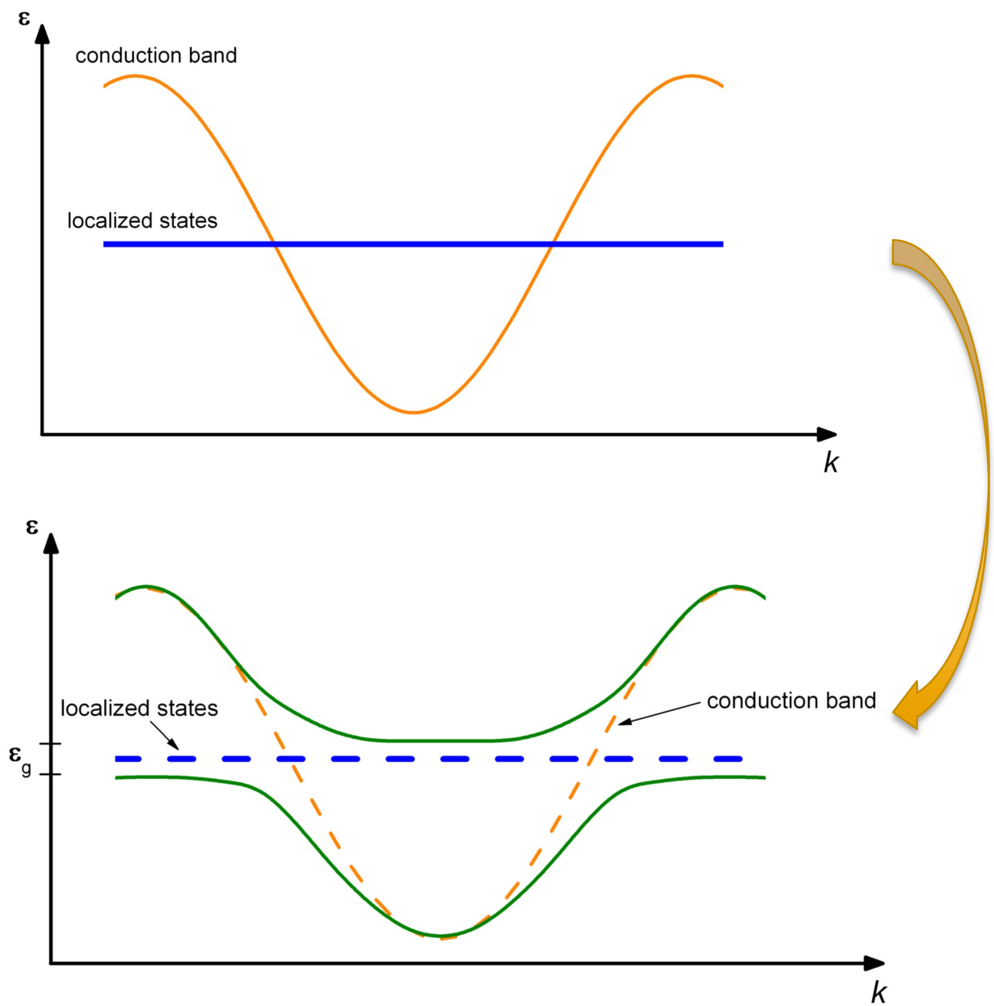
Kondo insulators

- **Kondo insulators a direct consequence of electronic correlations**

- Hybridization between the localized state and conduction electrons

- Chemical potential is located inside the hybridization gap

- Kondo insulators potentially topological

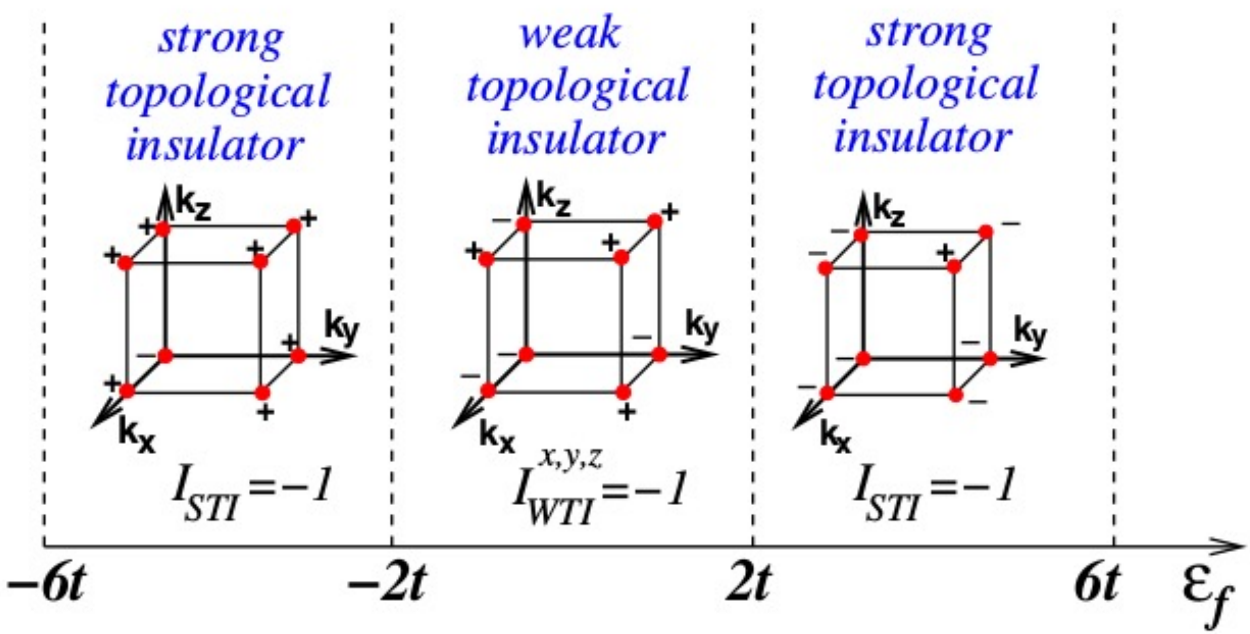


Topological Kondo insulators

- Periodic Anderson lattice Hamiltonian

$$\hat{H} = \sum_{\mathbf{k},\alpha} \xi_{\mathbf{k}} c^{\dagger}_{\mathbf{k}\alpha} c_{\mathbf{k}\alpha} + \sum_{j\alpha} [V c^{\dagger}_{j\alpha} f_{j\alpha} + \text{H.c.}]$$
$$+ \sum_{j\alpha} \left[\varepsilon_f^{(0)} n_{f,j\alpha} + \frac{U_f}{2} n_{f,j\alpha} n_{f,j\bar{\alpha}} \right]$$

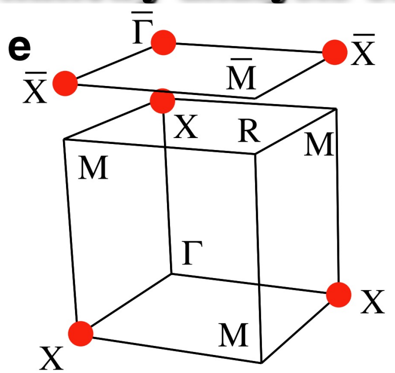
Parity eigenvalues of the high symmetry point is calculated:



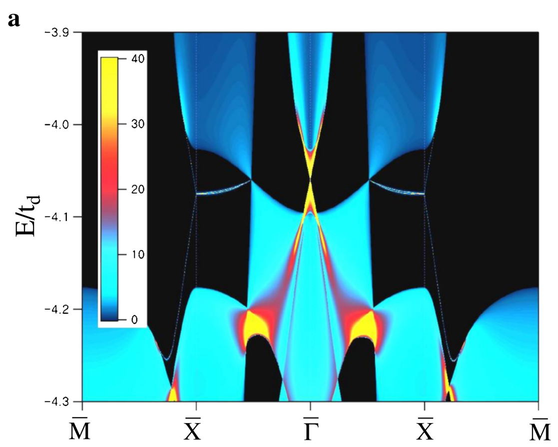
Dzero *et al.* PRL **104**, 106408 (2010)

TKI candidate: SmB_6

- **Symmetry analysis of the DFT band structure**

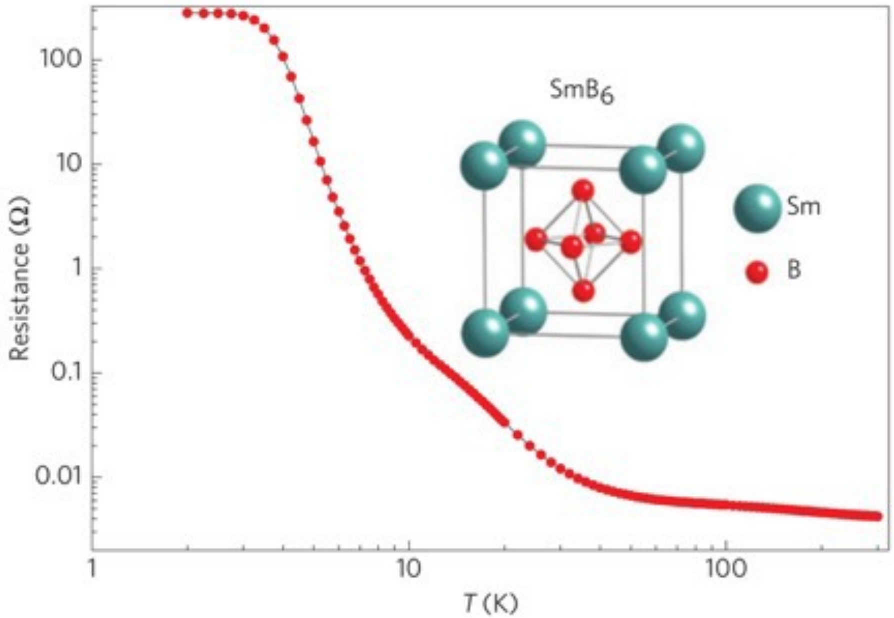


1. Strong topological insulator
(1; 1, 1, 1)



2. Slab calculation shows Dirac cones.

Takimoto, JPSJ **80**, 123710 (2011)



Kim et al., Nature Materials 3, 466 (2014)

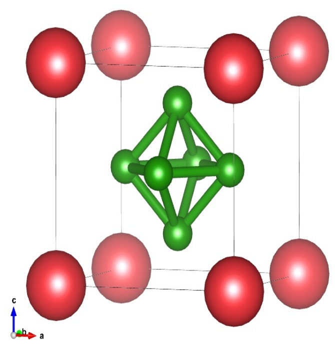
- Existence of surface states explains the saturation of resistivity in SmB_6

PuB$_6$ predicted TKI

- Cubic structure

a. Pu atoms are located at the corner.

b. 6 boron atoms form an octahedron at the center.

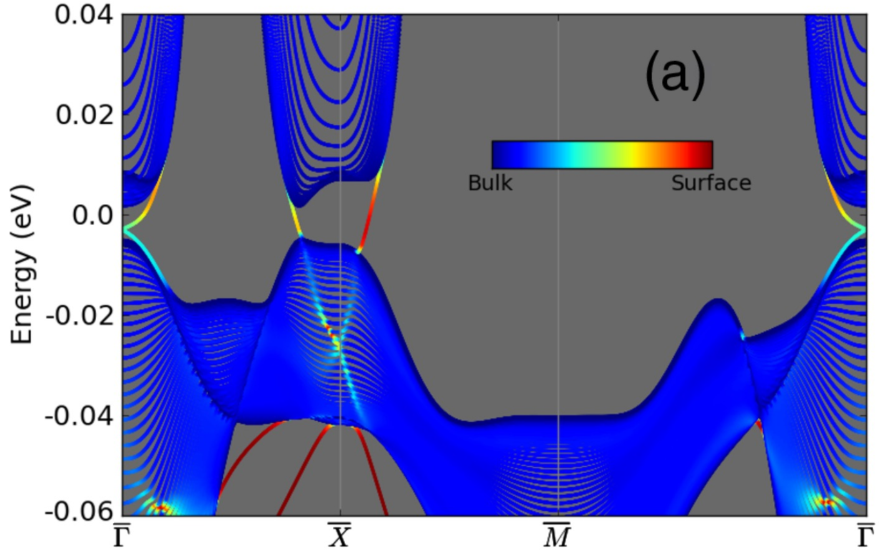


- Similarity to SmB$_6$

a. Same structure

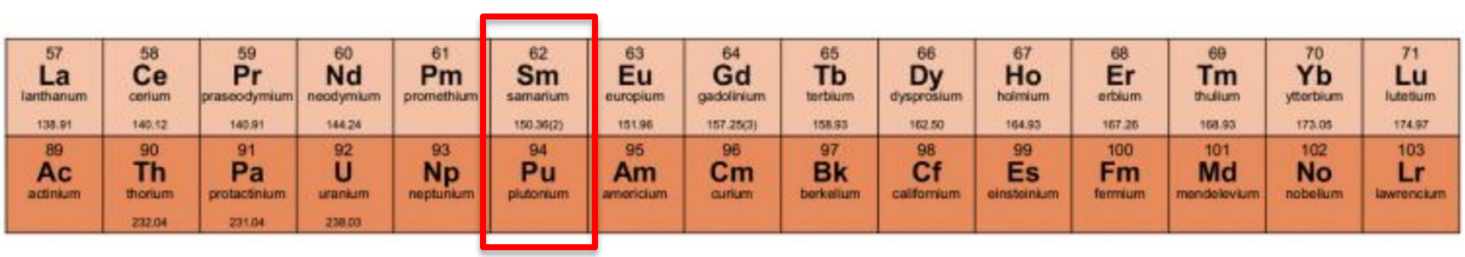
b. Same valence state is expected.

- Even stronger SOC



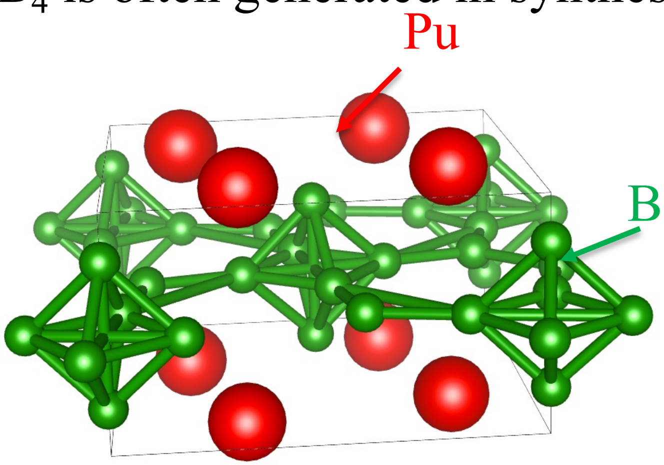
Deng *et al.*, PRL **111**,176404 (2013)

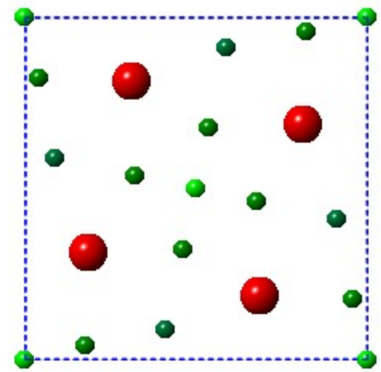
- Our goal – Search for 5*f*-electron topological Kondo insulators



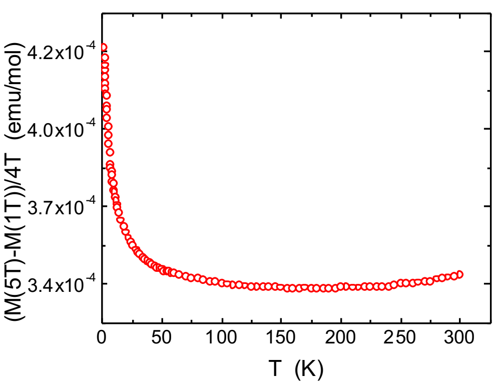
PuB_4 crystal structure

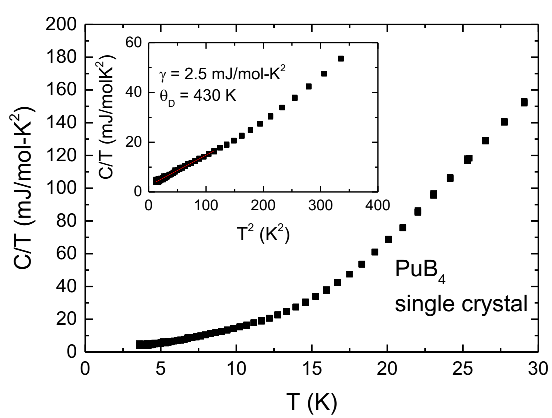
- Tetragonal structure (four formula units in the crystal unit cell)
- RB_4 is often generated in synthesis of RB_6.

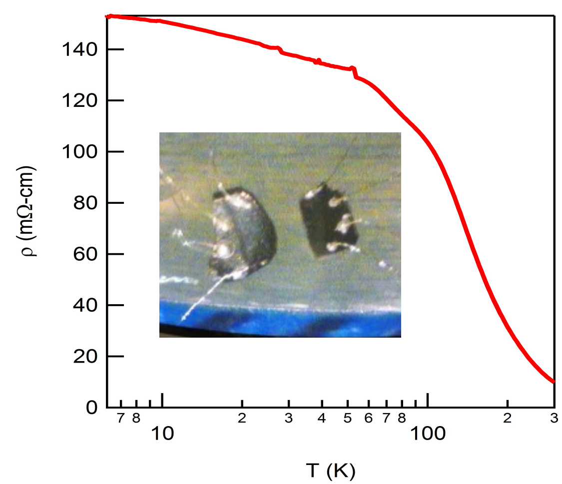




Experiment: Paramagnetic insulator with gap ~35 meV

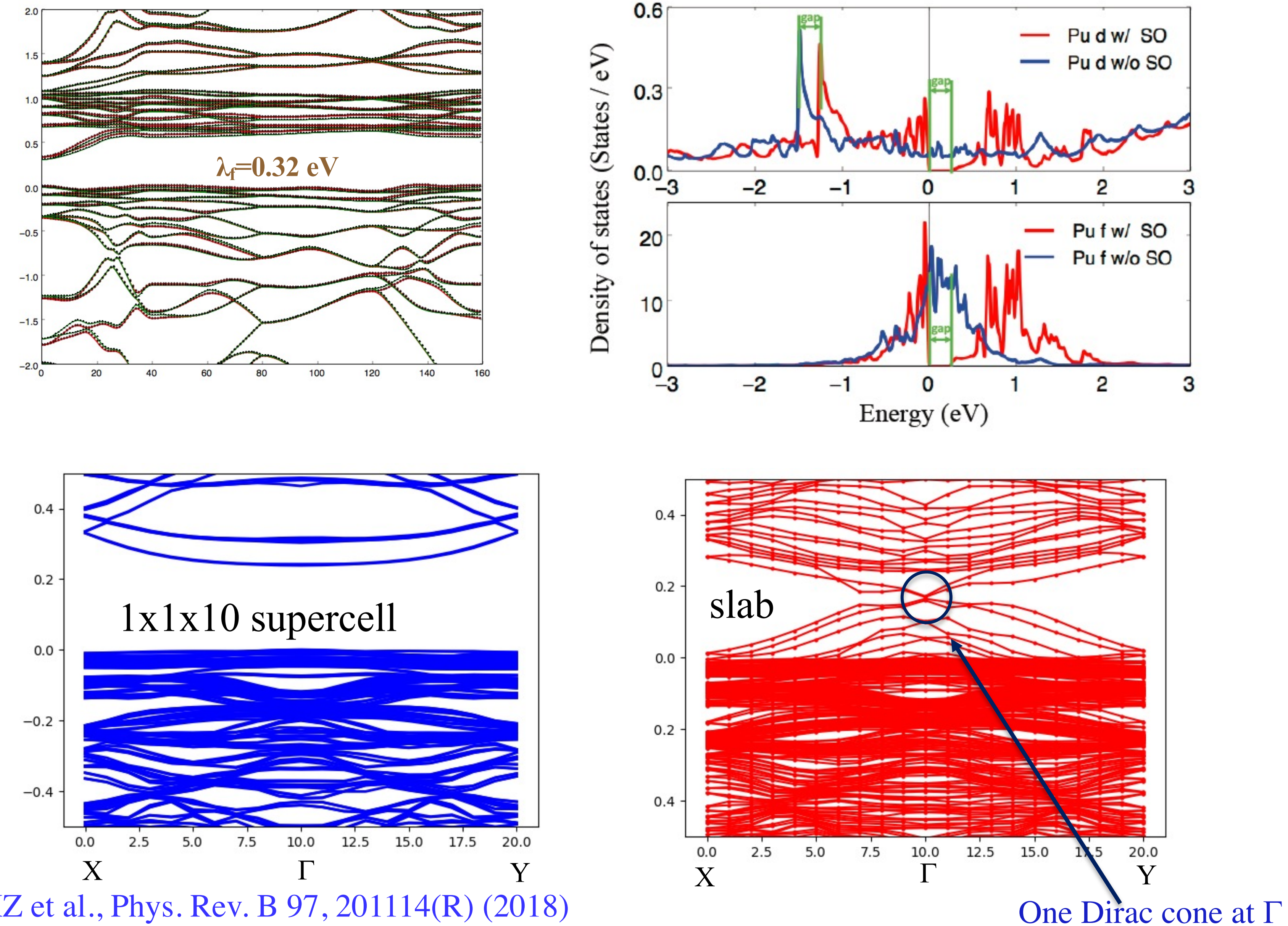






Choi, JXZ et al., Phys. Rev. B 97, 201114(R) (2018)

Spin-orbit coupling effect on PuB_4



Choi, JXZ et al., Phys. Rev. B 97, 201114(R) (2018)

Correlation effect on the gap

- In SmB6, it is understood that the strong electronic correlation renormalizes the theoretical band gap down to the experimental value without changing the topology.
- DFT+DMFT method: On-site Coulomb interaction (F^2, F^4, F^6 included) is added to the Pu-5*f* electrons.
    - ➢ Gives no moment, consistent with experiments in PuB_4.

Correlation effect on the gap size					
U (eV)	0	3.5	4.0	4.5	Experiment
gap size (meV)	253.6	15.2	13.2	10.3	35

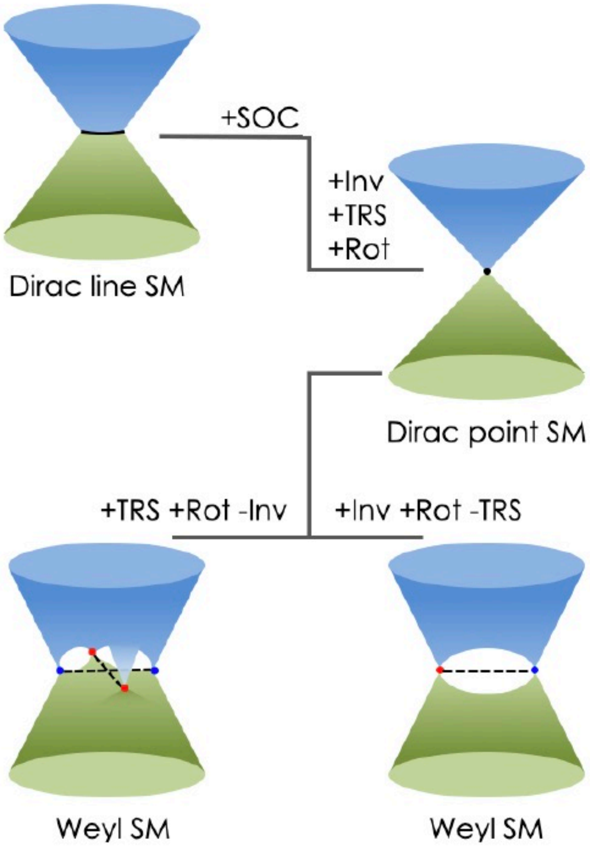
- ❖ The DFT+DMFT band gap is reduced by one order of magnitude relative to the overestimated DFT band gap. The correlation reduces the gap, supporting the notion that electronic correlations play an important role in PuB_4.
- ❖ Insulating phase is maintained throughout the entire range of U-strength, the robustness of the nontrivial topology is presumed.

Choi, JXZ et al., Phys. Rev. B 97, 201114(R) (2018)

Weyl semimetal?

- **Bulk states**:
  - Bands touch on gapless manifold
  - Fermi surface
- **Surface states**:
  - Fermi arcs, drumhead, etc.

- How to get topological metals?
  - Crystal symmetry → **degeneracy** $S^{-1}HS = H$
  - **Band inversion**: spin orbit coupling, strain, relativistic effects, etc.
  - Tune with **time-reversal** symmetry, **inversion** symmetry breaking

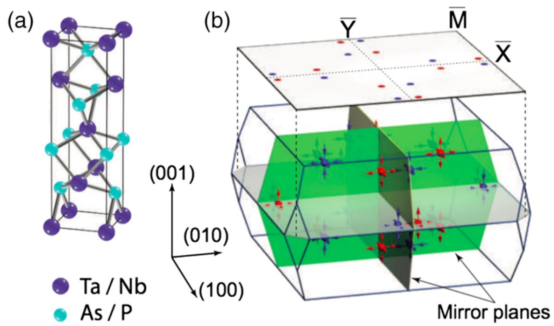


Yang et al., Adv. Phys.:X **3**, 1414631 (2018)

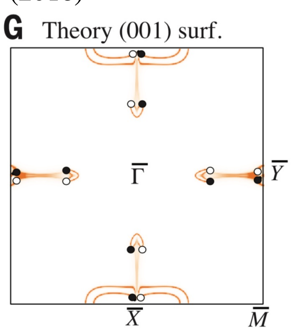
Material candidates for Weyl semimetals

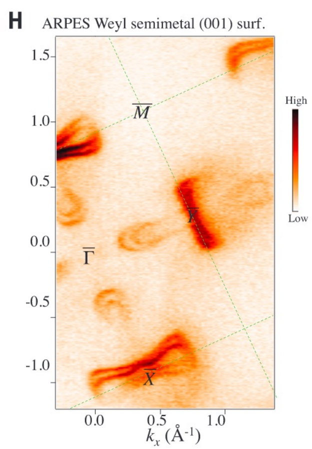
- Broken inversion symmetry

TaAs



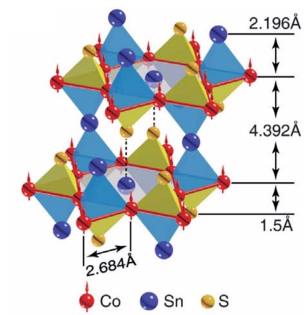
Weng et al. Phys. Rev. X 5, 011029 (2015)
Armitage et al. Rev. Mod. Phys. 90, 015001 (2018)

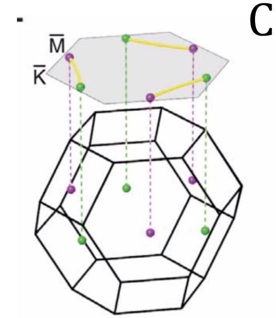




Xu et al. Science 349, 613 (2015)

- Broken time-reversal symmetry



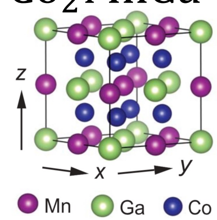


$Co_3Sn_2S_2$

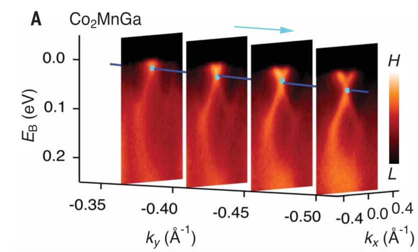
Morali et al., Science 365, 1286 (2019)

Liu et al., Science 365, 1282 (2019)

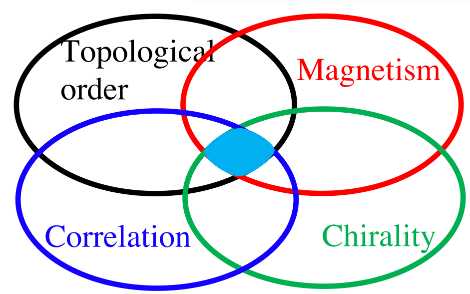
Co_2MnGa



Belopolski et al., Science 365, 1278 (2019)

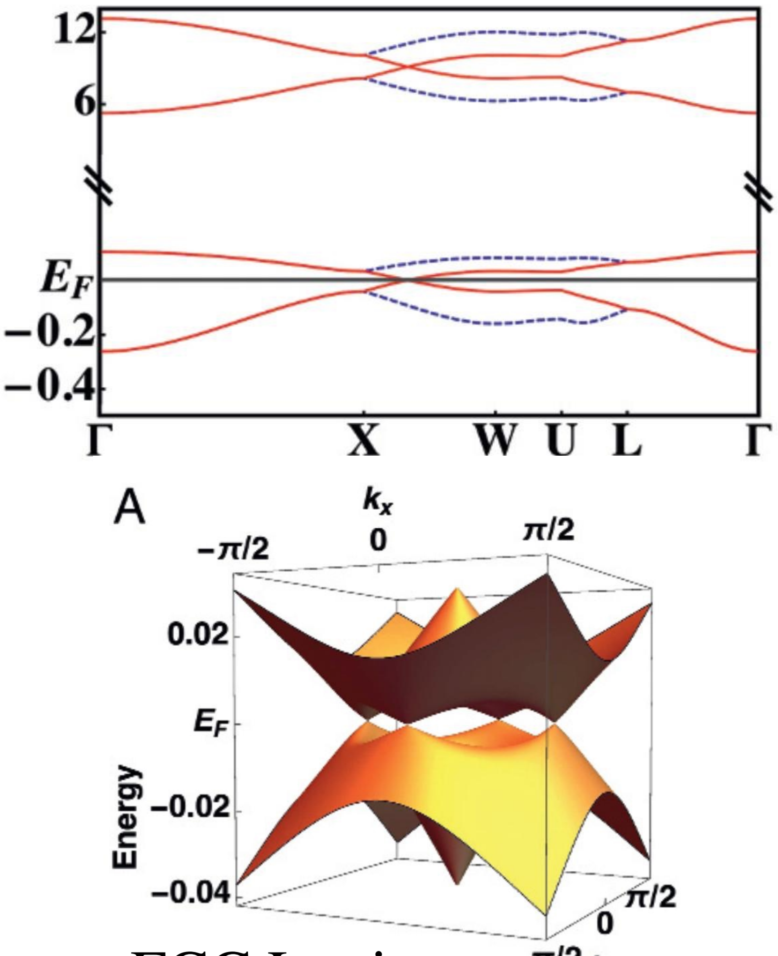


Weyl nodal line



far from enough to explore the wide range of possible exotic states

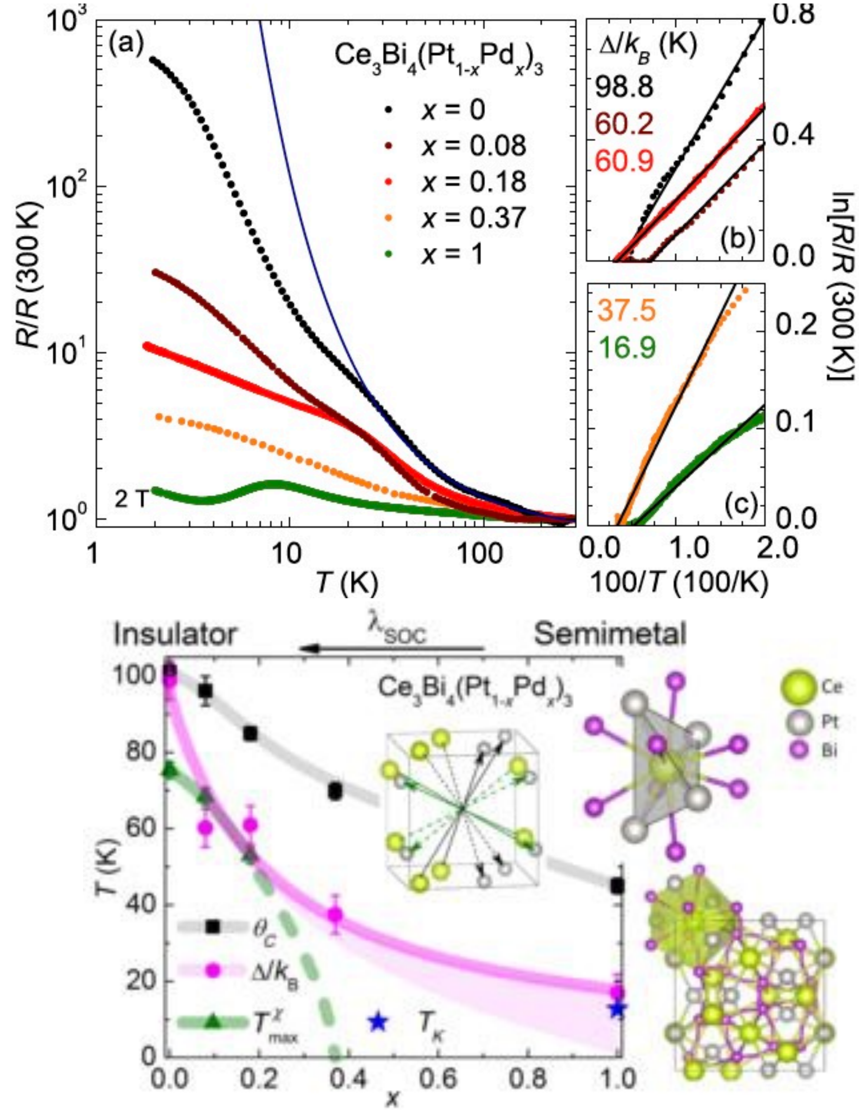
Candidates of other exotic state: Kondo Weyl semimetal?



FCC Lattice

Kondo Weyl Phase

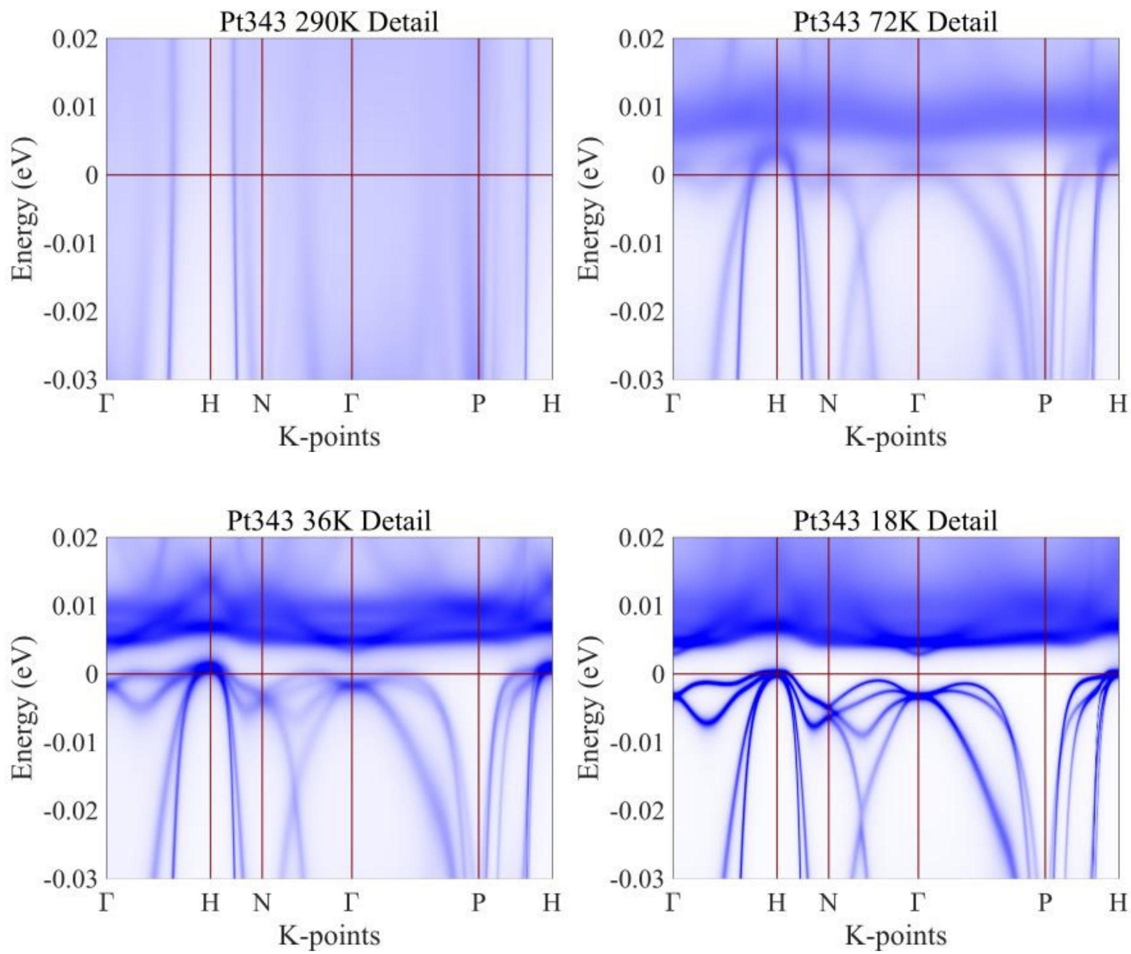
Lai et al., PNAS 115, 93



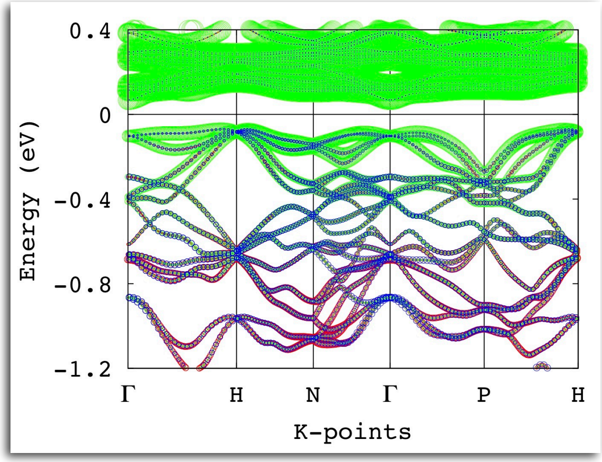
Dzsaber et al., PRL 118, 246601

Visualization of Kondo insulator gap opening in Pt334

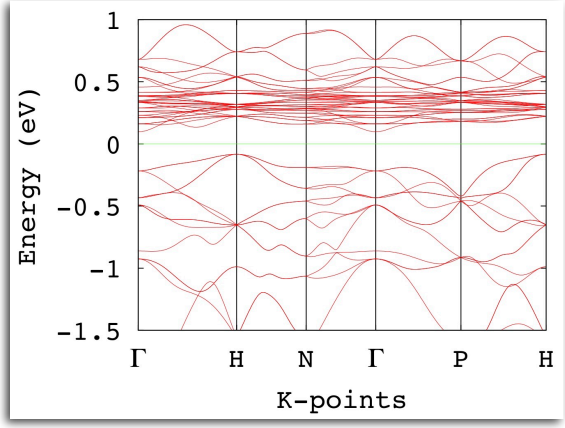
Kondo gap forming: 36 ~ 72 K
Δ ~ 6 meV @ 18K; $m^*/m=Z^{-1}$ ~ 26 @ 18K



DFT with SOC



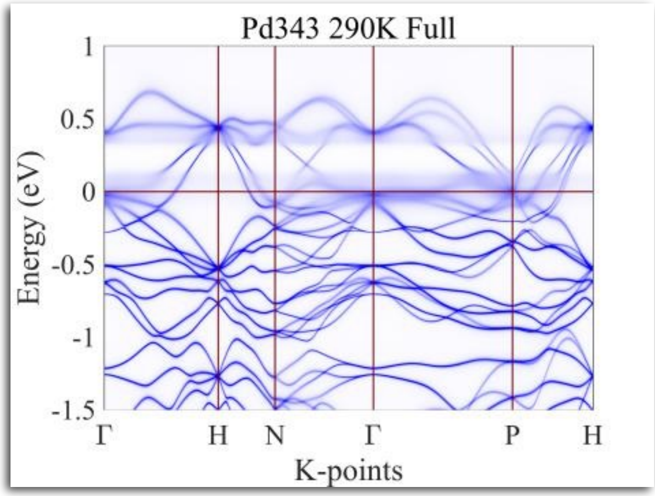
DFT without SOC

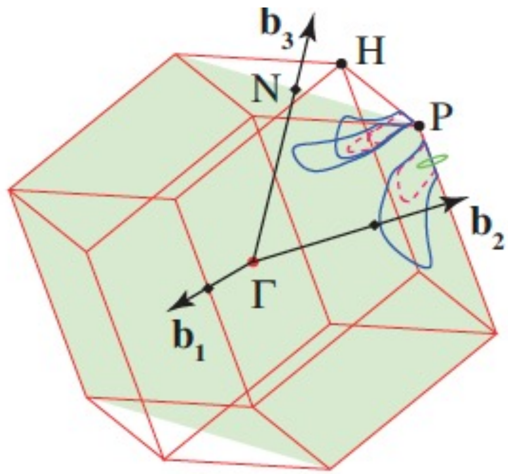


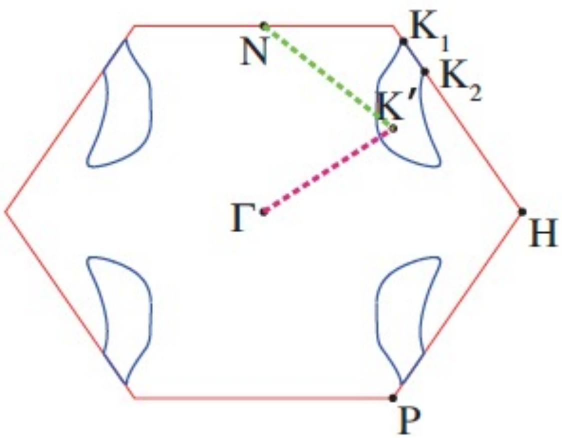
- **Pt334: trivial Kondo insulator**

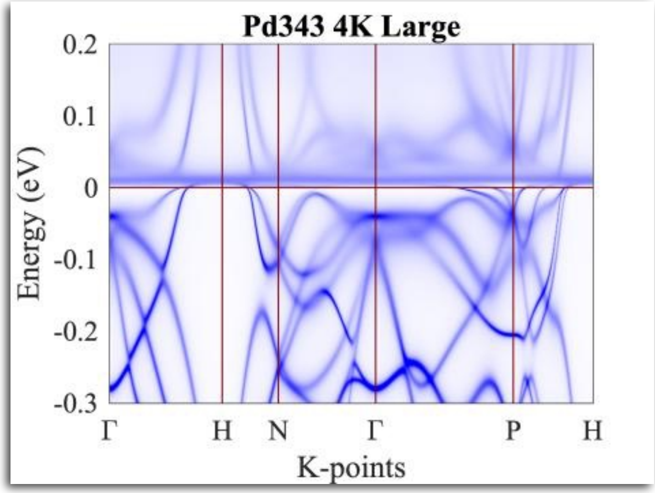
Cao, Zhi, JXZ, Phys. Rev. Lett. 124, 166403 (2020)

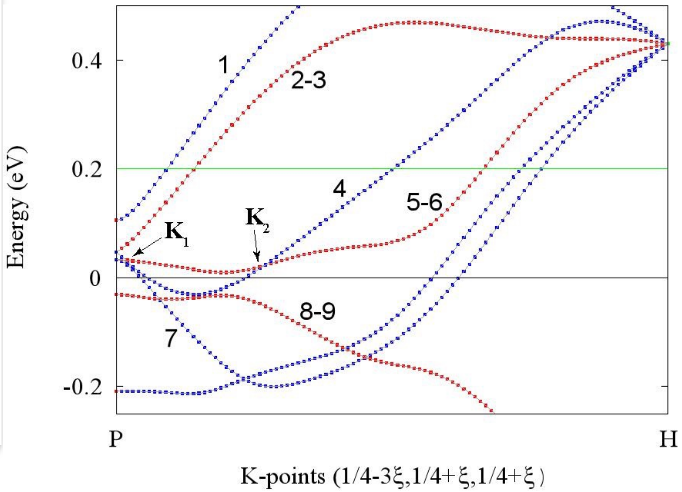
Temperature dependence of electronic structure in Pd334 (LDA+DMFT)









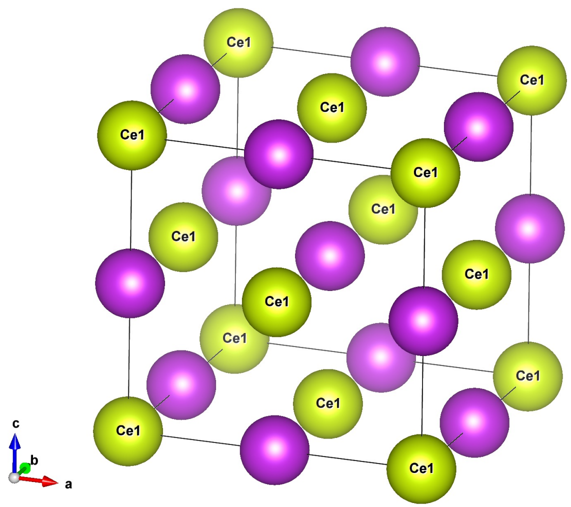


- Berry phase around the green solid line is π
- 3 nodal rings around each P point (eight P points in total) = 24 nodal rings in total

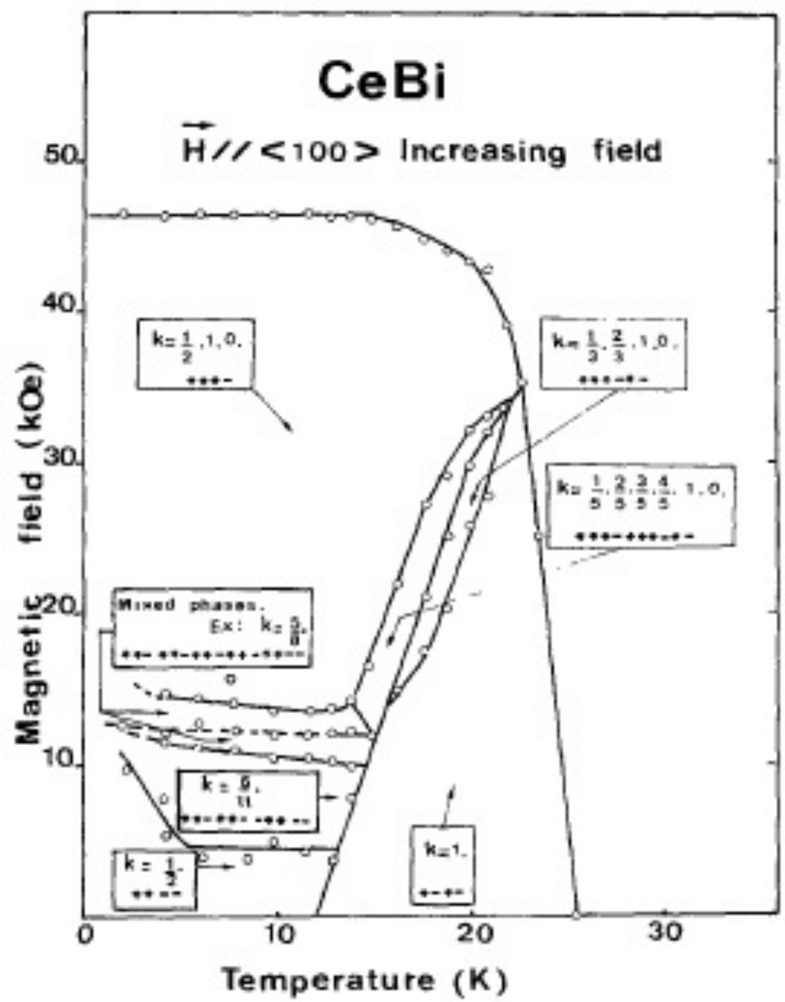
- **No hybridization gap down to 4K**
- **Ce-4f never coherent; coherent ligand states remain coherent**

Cao, Zhi, JXZ, Phys. Rev. Lett. 124, 166403 (2020)

CeBi: A magnetic Weyl semimetal



CeBi (H, T) phase dirgram

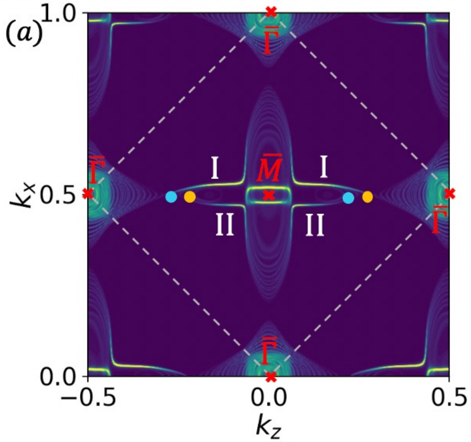


Bartholin et al., J. Physique, C5-30 (1979).

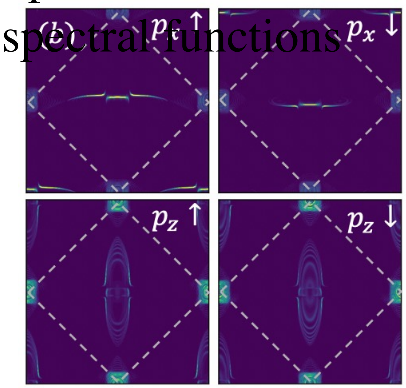
Fermi arcs on (010)-oriented surface

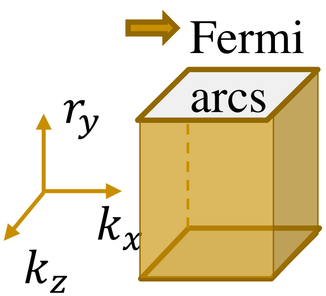
$E_{w_1} = 15\text{meV}$

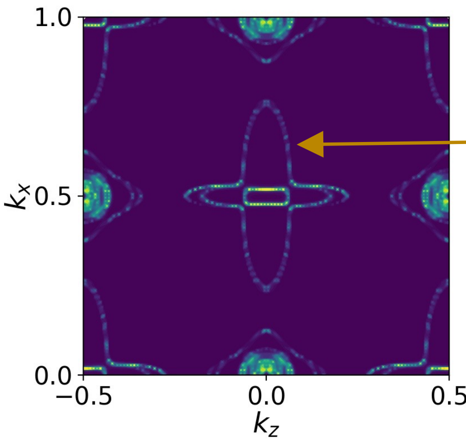
❑ Surface spectral function



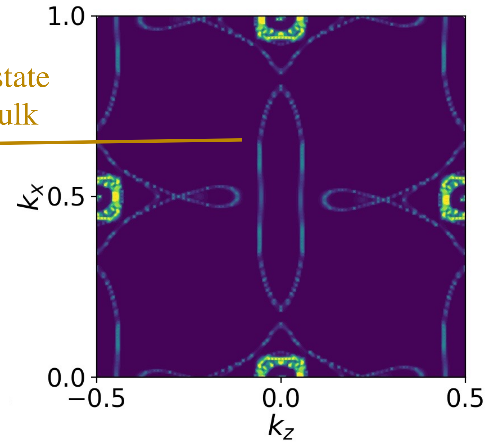
❑ Spin- and orbital-resolved surface spectral functions



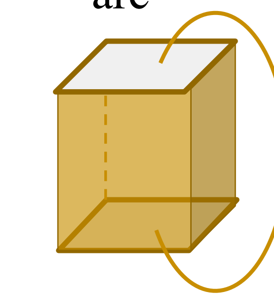




Trivial surface state derived from bulk band



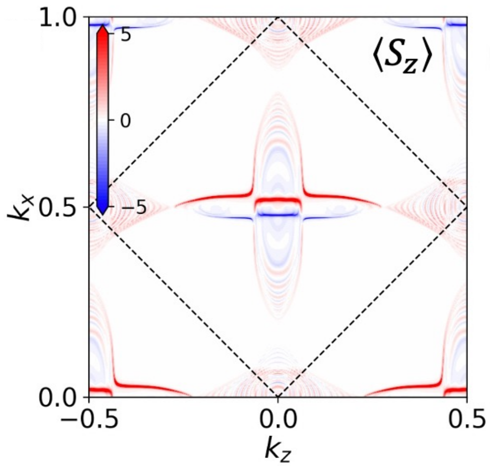
No Fermi arc



Huang, JXZ et al., Phys. Rev. B102, 235167 (2020)

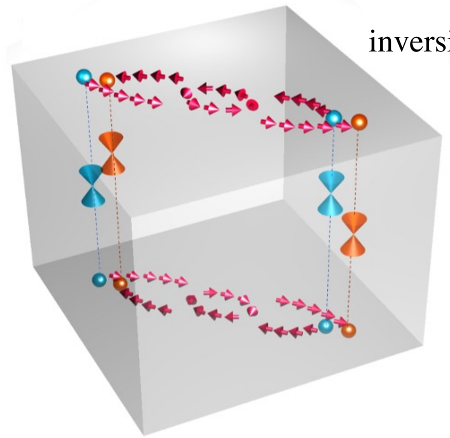
Spin texture of (010)-oriented surface

- σ_z projection of surface spectral function



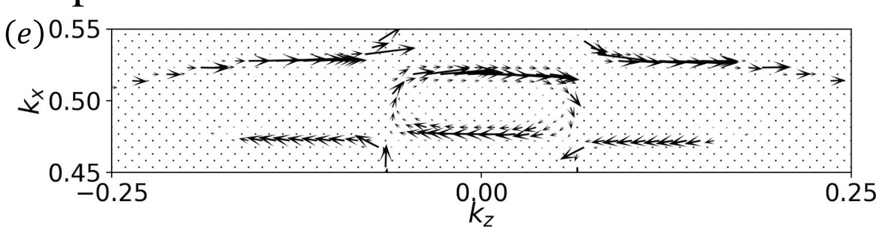
$$\langle S_i \rangle = -\frac{1}{\pi}\mathrm{ImTr}[\sigma_i G_{0s}]$$

- Schematic of spin texture for top and bottom surface in Brillouin zone



Spin: only in-plane directions

- Spin texture around the Fermi arcs from



Spin-polarized Fermi arcs and a spin vortex

- An important symmetry: $C_{2y}T$

$$C_{2y}TH(k_z,k_x)T^{-1}C_{2y}^{-1} = H(k_z,k_x)$$

$(C_{2y}T)^2 = 1$ No Kramers degeneracy

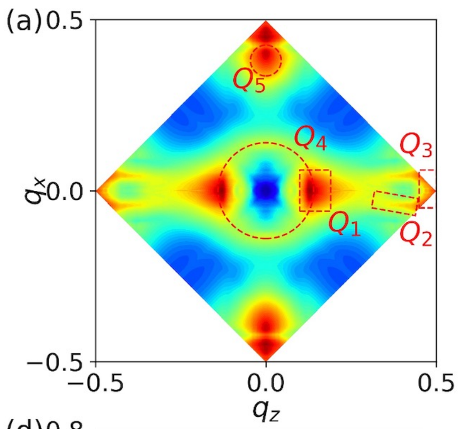
$C_{2y}T|k_z,k_x\rangle$ and $|k_z,k_x\rangle$ can only differ by a phase

$C_{2y}T|k_z,k_x,\uparrow_y\rangle = |k_z,k_x,\downarrow_y\rangle \Rightarrow \langle S_y\rangle$ must be zero

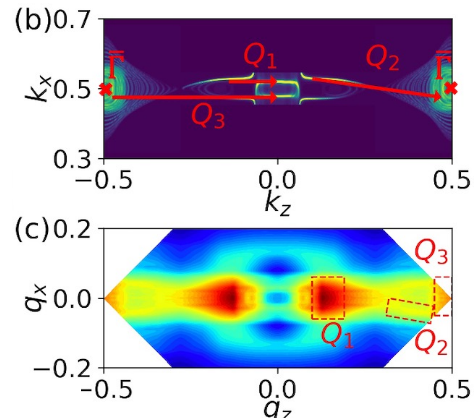
Huang, JXZ et al., Phys. Rev. B102, 235167 (2020)

Quasiparticle interference of (010)-oriented surface

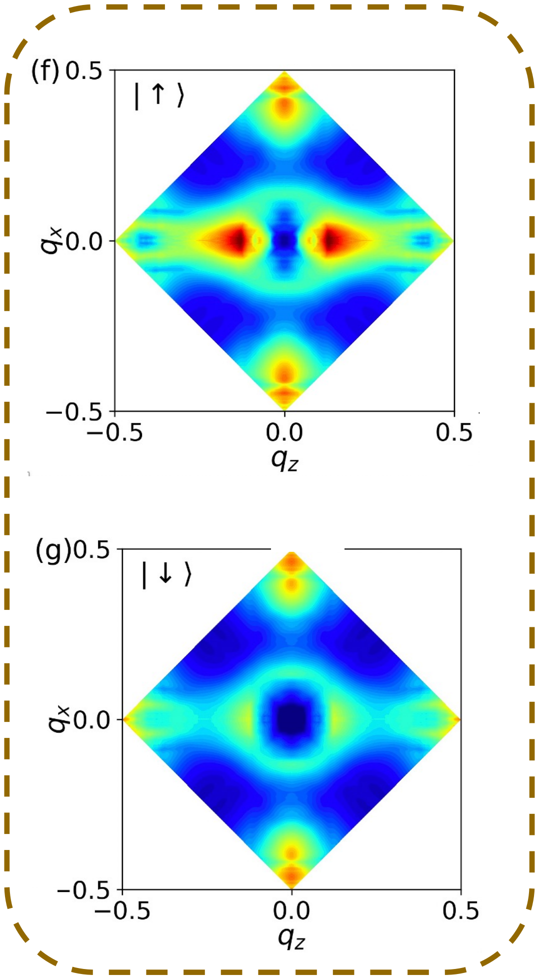
- QPI spectra with five main features



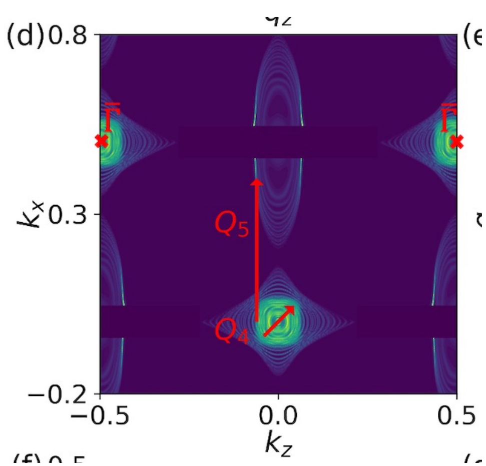
- scattering between the Fermi arcs, spin vortex, and bands around $\bar{\Gamma}$

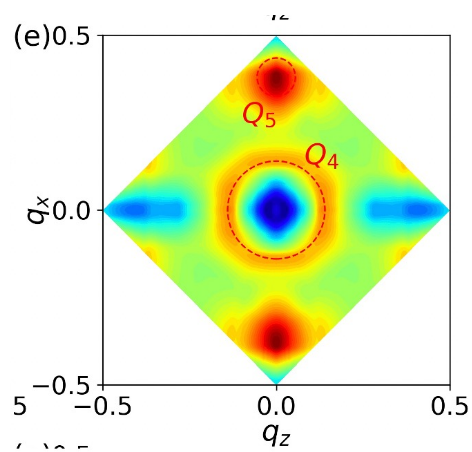


dependent QPI



- Scattering between regions away from the Fermi arcs and spin vortex





Huang, JXZ et al., Phys. Rev. B102, 235167 (2020)

Summary

- PuB_4 is a strong topological insulator.
  - PuB_4 shares many similarities with SmB_6, but with a larger energy scale.
  - Correlation effects are important for gap renormalization
- Topology in Ce-based 4f-electron systems
  - Pt334 is a trivial Kondo insulator
  - Pd334 is a nodal-line Kondo semimetal
  - Hybridization (Pt-5*d* vs Pd-4*d*) tuning is dominant mechanism
- CeBi is a magnetic Weyl semimetal
  - Fermi arc signature in ARPES and STM

f-electron quantum materials provides a powerful material platform to explore exotic states from the interplay of electronic correlation and topology

Acknowledgments

Collaborators

Theory

Christopher Lane, Zhao Huang (LANL)

Hongchul Choi (Seoul National Univ)

Wei Zhu (Westlake IAS)

Chao Cao (Hangzhou Normal Univ)

Experiment

Eric Bauer, Filip Ronning (LANL)

Dmitry Yarotski, Toni Taylor (LANL)

Yu Liu, Christian Matt (Harvard)

Brinda Kuthanazhi, Paul Canfield (Ames Lab)

Funding Support
LANL LDRD-DR & DOE BES

LDRD
innovation for our nation

- Dirac equation for spin-½ particle (yr. 1928)

❖ Existence of antiparticles

$$\left[\gamma^{\mu} p_{\mu} + m\right]\Psi = 0,$$

$$\gamma^0 = \begin{pmatrix} I & 0 \\ 0 & -I \end{pmatrix}, \gamma^i = \begin{pmatrix} 0 & \boldsymbol{\sigma} \\ -\boldsymbol{\sigma} & 0 \end{pmatrix}$$

Pauli Dirac

- Weyl equation for massless spin-1/2 particles (yr. 1929)

❖ Massless fermions with chirality

$$\left[\gamma^{\mu} p_{\mu} - m\right]\Psi = 0,\ p_{\mu} = \left(E, -\boldsymbol{p}\right) \Rightarrow \boldsymbol{\sigma} \bullet \boldsymbol{p}\psi_R = E\psi_R,$$

$$\gamma^0 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \gamma^i = \begin{pmatrix} 0 & -\sigma^i \\ \sigma^i & 0 \end{pmatrix} \qquad -\boldsymbol{\sigma} \bullet \boldsymbol{p}\psi_L = E\psi_L,$$

Hermann Weyl

- Majorana equation for massless spin-1/2 particles (yr. 1937)

❖ Fermion its own antiparticle

$$\left[-i\gamma^{\mu} p_{\mu} + m\right]\Psi = 0,$$

$$\gamma^{0,2} = \begin{pmatrix} 0 & \pm\sigma_2 \\ \sigma_2 & 0 \end{pmatrix}, \gamma^1 = \begin{pmatrix} i\sigma_3 & 0 \\ 0 & i\sigma_3 \end{pmatrix}, \gamma^3 = \begin{pmatrix} -i\sigma_1 & 0 \\ 0 & -i\sigma_1 \end{pmatrix}.$$

Ettore Majorana